Underbarrier nucleation kinetics in a metastable quantum liquid near the spinodal

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We develop a theory in order to describe the effect of relaxation in a condensed medium upon the quantum decay of a metastable liquid near the spinodal at low temperatures. We find that both the regime and the rate of quantum nucleation strongly depend on the relaxation time and its temperature behavior. The quantum nucleation rate slows down with the decrease of the relaxation time. We also discuss the low temperature experiments on cavitation in normal ³He and superfluid ⁴He at negative pressures. It is the sharp distinctions in the high frequency sound mode and in the temperature behavior of the relaxation time that make the quantum cavitation kinetics in ³He and ⁴He completely different in kind.

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1. INTRODUCTION

The phenomenon of macroscopic quantum nucleation in a metastable condensed medium, which is one of possible examples of the macroscopic quantum tunneling phenomena, has attracted a noticeable attention of researchers during recent years [1, 2]. The range of the metastable systems in which the macroscopic quantum nucleation is currently investigated is rather wide. These are from the helium systems involving, in particular, solidification from the overpressurized liquid phase [3], various types of sound-induced nucleation [4, 5], phase separation of supersaturated ³He-⁴He superfluid mixtures [6], and cavitation of bubbles at negative pressures [7, 8] to the collapse of the Bose-Einstein condensate in a Bose gas with attraction [9] and the formation of a quark matter in the core of neutron stars [10].

The modern representation on the decay of a metastable condensed medium is associated, first of all, with the nucleation of a seed of the stable phase and with the concept of a critical nucleus sufficient to overcome some potential barrier due to either thermal or quantum fluctuations and then to convert the whole metastable phase into the stable one. The decay rate due to its exponential behavior as a function of the barrier height depends drastically on the imbalance between the phases. As a rule, it is believed that the experimentally observable decay rates should be connected with the vicinity of a metastable medium to some instability resulting in the corresponding reduction of a potential barrier which prevents from the formation of a critical nucleus. In particular, in the case of the homogeneous cavitation of gas bubbles in a liquid such instability can be associated with approaching to the spinodal line for the liquid-vapor phase transition. The latter is one of the fundamental conclusions in the recent experiments on the quantum cavitation of bubbles in liquid ³He and ⁴He at negative pressures [11]. The next

important aspect of the experimental observations is a crossover in the temperature behavior of the attainable cavitation pressure. This is treated as an evidence for the thermal-quantum crossover in the cavitation kinetics.

Unfortunately, so far the quantum decay of a metastable condensed medium close to instability has not received an appropriate attention with the exception of works [12, 13] in which any relaxation processes in a medium are completely ignored in consideration. However, nobody can deny an importance of involving relaxation processes especially for the case of quantum liquids due to strong effect of temperature on the relaxation time in quantum media at low temperatures. Here we eliminate the shortages of the previous theory and involve the effect of relaxation processes accompanied by irreversible energy dissipation upon the underbarrier nucleation kinetics.

2. ENERGY AND DISPERSION SPECTRUM OF DENSITY FLUCTUATIONS

It is long known that the critical nuclei responsible for the decay of a metastable condensed medium near the spinodal are characterized by the relatively smooth boundaries and more extended sizes compared with the case of nucleation near the binodal corresponding to thermodynamic equilibrium between the phases. That is why, the van der Waals approach or gradient decomposition is usually employed for the description of fluctuations in a medium. Accordingly, the energy of a reversible fluctuation in a liquid reads as [12, 14]

$$U[\delta\rho] = \int d^3r \left[\frac{\varepsilon''(\rho)}{2} (\delta\rho)^2 + \frac{\varepsilon'''(\rho)}{6} (\delta\rho)^3 + \frac{\lambda(\rho)}{2} (\nabla\delta\rho)^2 + \ldots \right]$$
(1)

where ρ is the initial density of the homogeneous metastable medium close to the density ρ_c at the spinodal and $\delta \rho = \delta \rho(\mathbf{r}, t)$ is the fluctuative deviation of the density. The second derivative of the energy per unit volume

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of a liquid can be expressed via either compressibility $(\rho^2 \epsilon''(\rho))^{-1}$ or sound velocity $c_0(\rho)$

$$\epsilon''(\rho) = c_0^2(\rho)/\rho$$

The spinodal, associated with the violation of thermodynamic inequality $c_0^2(\rho) > 0$ and absolute instability against longwave fluctuations [15], is determined by vanishing the sound velocity, i.e.,

$$c_0(\rho_c) = 0 \tag{2}$$

The parameter λ , depending in general on density, determines a scale of the energy of inhomogeneity and can be interpreted in terms of dispersion of the sound spectrum as a function of wave vector. Though the energy of inhomogeneity also contributes into the interfacial tension usually measured at the binodal, one should treat this very carefully. In fact, in this case one may need to involve next orders into the gradient expansion since the thickness of the interface at the binodal does not much exceed several interatomic distances and the change in density across it is noticeable with the exception of the immediate vicinity of the liquid-vapor critical point.

Involving condition (2), we can represent the potential energy of a density fluctuation (1) as

$$U[\delta\rho] = \int d^3r \left[\frac{c_0^2(\rho)}{2\rho} \left(\delta\rho^2 + \frac{1}{3} \frac{\delta\rho^3}{\rho - \rho_c} \right) + \frac{\lambda}{2} (\nabla\delta\rho)^2 + \dots \right]$$
(3)

The restriction with cubic terms in the expansion implies that the sound velocity vanishes as $c(\rho) \propto (\rho - \rho_c)^{1/2}$ or $c(P) \propto (P - P_c)^{1/4}$ as a function of pressure in the vicinity of the spinodal pressure P_c corresponding to density ρ_c . At present, the genuine exponent in the behavior of the sound velocity in liquid helium near the spinodal is unknown. Also, it is frequently proposed that $c(P) \propto (P - P_c)^{1/3}$ and $c(\rho) \propto (\rho - \rho_c)$. This estimate is based mainly on extrapolating the sound velocity data from the range of positive pressures to negative ones [16]. An explanation of such wholly satisfactory extrapolation beyond the close vicinity of the spinodal point can be found, e.g., in [17]. To describe the latter behavior which is more typical for the vicinity of the liquid-vapor critical point, we must retain the terms of expansion to fourth order in $\delta \rho$. Though this case requires a special treatment, we believe that the qualitative picture of nucleation remains faithful with the exception of numerical coefficients in the final expressions.

Provided we are interested only in thermal fluctuations, Eq. (1) or Eq. (3) is, in principle, sufficient to determine the decay rate within the exponential accuracy since the nucleation process is mainly governed by the saddle point of the functional of potential energy. In order to investigate the underbarrier nucleation kinetics and to calculate the rate of quantum decay, we employ the formalism in terms of imaginary-time path integrals and based on the use of the finite-action solutions (instantons) of equations of motion continued to the imaginary time. For review, see, e.g., Refs. [1, 2]. This approach of the effective Euclidean action was used for describing quantum decay of a metastable condensed medium near the binodal in order to incorporate the energy dissipation effect on the quantum kinetics of first-order phase transitions at low temperatures [18, 19].

Any fluctuation as a perturbation violates the thermodynamic equilibrium in the liquid, triggering the internal processes to recover the equilibrium. Small oscillations of the density represent a superposition of sound waves. The character of sound processes depends strongly on a ratio between the typical inverse frequency of density fluctuations and the typical time of relaxation processes in a liquid. The finiteness of the relaxation times results, in particular, in the frequency dispersion of the sound velocity and in the sound attenuation. Obviously, this effect is important for quantum liquids in which the relaxation times depend significantly on temperature.

In general, deriving the exact equation of sound dispersion for the whole range of frequencies is practically unsolvable problem. Usually, in order to describe the experimental observations, the so-called τ -approximation of a single relaxation time is employed. Then, the dispersion equation, i.e., relation between wave vector k and frequency ω , reads, e.g., [20]

$$c_0^2 k^2 = \omega^2 \frac{1 - i\omega\tau}{1 - i\omega\tau(c_\infty/c_0)^2} \tag{4}$$

Here $\tau = \tau(T)$ is a relaxation time depending, in general, on temperature T. The low frequency $\omega \tau \ll 1$ limit corresponds fully to the usual hydrodynamic sound with attenuation coefficient

$$\gamma(\omega) = \frac{\omega^2 \tau}{2c_0} \left(\frac{c_\infty^2}{c_0^2} - 1 \right)$$

which can be expressed in terms of viscosity $\eta=(3/4)\rho c_0^2\tau(c_\infty^2/c_0^2-1)$. The sound velocity $c_\infty>c_0$ stands for the velocity of high frequency $\omega\tau\gg 1$ collisionless sound with the attenuation coefficient proportional to $1/\tau$. In liquid ³He the high frequency limit can be associated with the zero-sound mode and Eq. (4) approximates well both the frequency dispersion of sound velocity and the attenuation within the accuracy of several percent.

3. QUANTUM DESCRIPTION AND EFFECTIVE ACTION

Let us return to the underbarrier nucleation. The probability of the quantum decay will be proportional to

$$W \propto \exp(-S/\hbar)$$

where S is the effective Euclidean action taken for the optimum path from the entrance point under the potential

barrier to the point at which the optimum fluctuation escapes from the barrier. So, for calculating quantum probability, we must construct the effective action determined in the imaginary time so that the energy (3) of a fluctuation would play a role of a potential energy. In addition, in accordance with the principle of analytic continuation into the real time the extremum path obtained by varying the effective action and analytically continued from the imaginary Matsubara frequencies to real ones ($|\omega_n| \rightarrow -i\omega$) should reproduce the classical equation of motion, i.e., dispersion Eq.(4) which the dynamics of small density fluctuations obeys in the real time.

Due to small variations of the density for fluctuations near the spinodal we can describe the motion of a liquid with the aid of the field of displacement $\boldsymbol{u}(\boldsymbol{r},t)$. In other words, we will treat the liquid phase as an elastic medium with zero shear modulus. A relative variation of the density is related to the change of the bulk element due to deformation

$$\frac{\delta\rho}{\rho} = \frac{\rho' - \rho}{\rho} = -\frac{dV' - dV}{dV'}$$

The bulk elements after and before deformation are connected with the Jacobian according to [21]

$$dV' = \parallel \delta_{ik} + \partial u_i / \partial r_k \parallel dV$$

where $\boldsymbol{u}(\boldsymbol{r},t)$ is the displacement vector describing the deformation of a medium.

Emphasize that we should employ the relation between density variation $\delta \rho$ and displacement $\boldsymbol{u}(\boldsymbol{r},t)$ beyond the linear approximation since cubic terms in $\delta \rho$ are involved into the expansion of the energy of a fluctuation. Second order in $\boldsymbol{u}(\boldsymbol{r},t)$ is sufficient for our purposes

$$\frac{\delta \rho}{\rho} \approx -\nabla \cdot \boldsymbol{u} + \frac{1}{2} \left[\left(\frac{\partial u_l}{\partial r_l} \right)^2 + \frac{\partial u_i}{\partial r_k} \frac{\partial u_k}{\partial r_i} \right]$$
 (5)

As a result, we arrive at the following nonlocal effective action

$$S[\boldsymbol{u}(\boldsymbol{r},t)] = \int_{-1/2T}^{1/2T} dt dt' \int d^3r \, \frac{\rho}{2} \dot{\boldsymbol{u}}(\boldsymbol{r},t) D(t-t') \dot{\boldsymbol{u}}(\boldsymbol{r},t') + \int_{-1/2T}^{1/2T} dt \int d^3r \, \left[\frac{c_0^2(\rho)}{2\rho} \left(\delta \rho^2 + \frac{1}{3} \frac{\delta \rho^3}{\rho - \rho_c} \right) + \frac{\lambda}{2} (\nabla \delta \rho)^2 \right]$$
(6)

Here we put $\hbar = 1$ and $k_B = 1$. The nonlocal kernel D(t) describing relaxation of a density fluctuation is simply

expressed in terms of its Fourier transform $D(\omega_n)$

$$D(t) = T \sum_{n} D(\omega_{n}) e^{-i\omega_{n}t} , \ \omega_{n} = 2\pi n T , \ n = 0, \pm 1, \pm 2, \dots$$

$$D(\omega_{n}) = \frac{1 + |\omega_{n}| \tau}{1 + |\omega_{n}| \tau c_{\infty}^{2}/c_{0}^{2}} = \begin{cases} 1 - |\omega_{n}| \tau \frac{c_{\infty}^{2} - c_{0}^{2}}{c_{0}^{2}}, & \tau c_{\infty}^{2}/c_{0}^{2} \ll |\omega_{n}|^{-1} \\ \frac{1}{|\omega_{n}|\tau} \frac{c_{0}^{2}}{c_{\infty}^{2}}, & \tau \ll |\omega_{n}|^{-1} \ll \tau c_{\infty}^{2}/c_{0}^{2}, \text{ if } c_{\infty} \gg c_{0} \\ \frac{c_{0}^{2}}{c_{\infty}^{2}} \left(1 + \frac{c_{\infty}^{2} - c_{0}}{c_{\infty}^{2}} \frac{1}{|\omega_{n}|\tau}\right), & |\omega_{n}|^{-1} \gg \tau \end{cases}$$

$$(7)$$

The latter is in the full correspondence with Eq. (4). The appearance of $|\omega_n|$ in (7) agrees with the general rule of analytic continuation $\omega \to i |\omega_n|$ for the Fourier transforms of retarded correlators in the course of transformation from the physical real time to imaginary time. Note that the case of zero relaxation time $\tau = 0$ corresponds to the action considered in [12] within the linear approximation $\delta \rho / \rho = -\nabla \cdot \boldsymbol{u}$ in (5). In [12] the nucleation dynamics was assumed to be completely reversible and thus any possible effect of relaxation and energy dissipation upon the underbarrier nucleation was ignored.

As a next step, it is convenient to introduce the dimen-

sionless units

$$\chi = \frac{1}{2} \frac{\delta \rho}{\rho - \rho_c} , \quad x = \frac{r}{l} , \quad \eta = \frac{t}{t_0} ,
\mathbf{v} = \frac{\mathbf{u}}{u_0} , \quad T' = Tt_0 , \quad \Omega_n = \omega_n t_0$$
(8)

Here we put

$$l^2 = \frac{\lambda \rho}{c_0^2(\rho)}$$
, $u_0 = 2l \frac{\rho - \rho_c}{\rho}$, and $t_0 = \frac{l}{c_0(\rho)}$

Accordingly, relation (5) goes over into

$$\chi = -\nabla \cdot \boldsymbol{v} + \frac{\rho - \rho_c}{\rho} \left[\left(\frac{\partial v_l}{\partial x_l} \right)^2 + \frac{\partial v_i}{\partial x_k} \frac{\partial v_k}{\partial x_i} \right]$$

and action (6) reduces to

$$S = S_0 s[\boldsymbol{v}(\boldsymbol{x}, \eta)]$$

where the dimensional factor is given by

$$S_0 = 4t_0 l^3 \frac{c_0^2(\rho)}{\rho} (\rho - \rho_c)^2 = 4\lambda^2 \rho \frac{(\rho - \rho_c)^2}{c_0^3(\rho)}$$
 (9)

The dimensionless action decomposed to third order in displacement \boldsymbol{v} reads

$$s[\boldsymbol{v}(\boldsymbol{x},\eta)] = \int_{-1/2T'}^{1/2T'} d\eta d\eta' \int d^3x \, \frac{1}{2} \dot{\boldsymbol{v}}(\boldsymbol{x},\eta) D(\eta - \eta') \dot{\boldsymbol{v}}(\boldsymbol{x},\eta')$$

$$+ \int_{-1/2T'}^{1/2T'} d\eta \int d^3x \, \left\{ \frac{1}{2} (\nabla(\nabla \cdot \boldsymbol{v}))^2 + \frac{1}{2} (\nabla \cdot \boldsymbol{v})^2 - \frac{1}{3} \left[(\nabla \cdot \boldsymbol{v})^3 - 3 \, \frac{\rho - \rho_c}{\rho} \left((\nabla \cdot \boldsymbol{v})^3 + \frac{\partial v_i}{\partial x_k} \frac{\partial v_k}{\partial x_i} \frac{\partial v_l}{\partial x_l} \right) \right] \right\}$$

$$(10)$$

where the Fourier transform of $D(\eta)$ is given by

$$D(\Omega_n) = \frac{1 + |\Omega_n| \tau/t_0}{1 + |\Omega_n| \tau c_{\infty}^2/t_0 c_0^2}$$

The reduction of the coefficient in the front of $(\nabla \cdot \boldsymbol{v})^3$ leads to increasing the extremum value of the action. However, keeping in mind the vicinity to the spinodal, we will neglect below the cubic terms proportional to $(\rho - \rho_c)/\rho \ll 1$. Note only that the involvement of these cubic terms yields a correction of the same order of $(\rho - \rho_c)/\rho$ into the final answer and can be treated as a perturbation.

4. QUANTUM NUCLEATION RATE

In this section we will analyze extremum paths of effective action (10) and, correspondingly, determine the nucleation rate. The exact determination of extrema is a rather complicated problem. We start from the limiting cases. First, we mention the time-independent path entailing the classical Arrhenius law for the nucleation rate. From (9) and (10) one readily obtains

$$S_{cl} = U_0/T$$

 $U_0 = 4(\lambda^3 \rho)^{1/2} \frac{(\rho - \rho_c)^2}{c_0(\rho)} f_0$

where U_0 plays a role of the potential barrier for nucleation and f_0 is the extremum saddle-point value of the functional for the potential energy given in dimensionless units

$$f[\chi(x)] = \int d^3x \left(\frac{1}{2}(\nabla \chi)^2 + \frac{1}{2}\chi^2 + \frac{1}{3}\chi^3\right)$$

The numerical solution for a critical fluctuation of the spherical symmetry results in $f_0 \approx 43.66$. (This value is

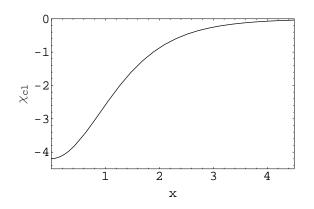
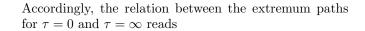


FIG. 1: The density profile of the thermal critical fluctuation.

about 9% larger than those used in [12, 13, 14].) The amplitude at the center is $\chi_{\rm cl}(x=0) \approx -4.19$ and becomes a half as much at the distance $x \approx 1.22$ [22]. The spatial behavior of critical fluctuation $\chi_{\rm cl}(x)$ is shown in Fig. 1.

Let us turn now to the case of zero temperature and consider the extremum value of effective action as a function of relaxation time τ . Action $s[\boldsymbol{v}(\boldsymbol{x},\eta] \ (10)$ on the neglect of the term proportional to $(\rho-\rho_c)/\rho$ was analyzed in [12] for the relaxation time $\tau=0$ at T=0. The extremum value $s_0(0)$ for a critical quantum fluctuation was estimated as $s_0(0)\approx 160$. The space-time behavior of the density for the quantum critical fluctuation at zero temperature is shown in Fig. 2.

The other limiting case, namely, infinite time of relaxation, can easily be reduced to the case of zero time by redefining factor ρ as $\rho_{\infty} = \rho c_0^2/c_{\infty}^2$ in the term with the kinetic energy in (6). This results in the exact relation between the cases $\tau = 0$ and $\tau = \infty$ since both S and T'



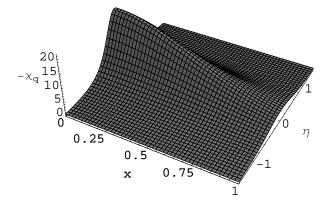


FIG. 2: The space-time profile of the density plotted with the minus sign, illustrating quantum critical fluctuation at zero temperature.

are proportional to $\sqrt{\rho}$

$$S_{\infty}(T) = \frac{c_0}{c_{\infty}} S_0 \left(\frac{c_0}{c_{\infty}} T \right)$$

$$\boldsymbol{v}_{\infty}(\boldsymbol{x},\,\eta) = \boldsymbol{v}_0(\boldsymbol{x},\,\eta c_{\infty}/c_0) \tag{11}$$

Employing the limiting expressions from (7) for kernel $D(\omega_n)$ and the relation

$$T\sum_{n} |\omega_n| e^{-i\omega_n t} = -\frac{\pi T^2}{\sin^2 \pi T t}$$
 (12)

we can estimate the effective action within first-order approximation of the theory of perturbations in two limiting cases of small $\tau \ll t_0 c_0^2/c_\infty^2$ and large $\tau \gg t_0 c_0/c_\infty$ relaxation time. For small relaxation times $\tau \to 0$, we find that correction $\Delta S_0(T)$ is negative and, therefore, finite time of relaxation facilitates nucleation of new phase as compared with the case of zero time of relaxation

$$S(T) = S_0(T) + \Delta S_0(T)$$

$$\frac{\Delta S_0}{S_0} = -\frac{c_\infty^2 - c_0^2}{\sqrt{\lambda \rho}} \tau \frac{1}{4\pi} \int_{-1/2T'}^{1/2T'} d\eta d\eta' \int d^3x \left(\dot{\boldsymbol{v}}_0(\boldsymbol{x}, \eta) - \dot{\boldsymbol{v}}_0(\boldsymbol{x}, \eta')\right)^2 \frac{(\pi T')^2}{\sin^2 \pi T'(\eta - \eta')}$$
(13)

where we used notation $\dot{\boldsymbol{v}}_0 = \partial \boldsymbol{v}_0/\partial \eta$. The other opposite

case $\tau^{-1} \to 0$ yields

$$S(T) = S_{\infty}(T) + \Delta S_{\infty}(T)$$

$$\frac{\Delta S_{\infty}}{S_{\infty}} = \frac{c_{\infty}^{2} - c_{0}^{2}}{c_{\infty}^{2}} \frac{\sqrt{\lambda \rho}}{c_{0}c_{\infty}} \frac{1}{\tau} \frac{1}{4\pi} \int_{-1/2T''}^{1/2T''} d\eta d\eta' \int d^{3}x \left(\mathbf{v}_{0}(\mathbf{x}, \eta) - \mathbf{v}_{0}(\mathbf{x}, \eta') \right)^{2} \frac{(\pi T'')^{2}}{\sin^{2}\pi T''(\eta - \eta')}$$
(14)

Here we involved Eq. (11) and denoted $T'' = T'c_0/c_{\infty}$. The sign of this correction is positive.

Concerning the intermediate case of relaxation time $t_0c_0^2/c_\infty^2 \ll \tau \ll t_0c_0/c_\infty$ which can be prominent only provided $c_\infty \gg c_0$, we should mention that the character of quantum nucleation in this case is different in kind from the two limiting ones considered above. The point is that nonlocal kernel $\Omega^2 D(\Omega)$ responsible for the tunneling process becomes effectively proportional to $|\Omega|$ instead of Ω^2 . Thus the dynamics of underbarrier nucle-

ation changes from the usual one governed by the kinetic energy term to the overdamped viscous type. The functional dependence on the physical parameters can readily be obtained by redefining time η from (8) as

$$\eta \to \frac{c_0^2}{c_\infty^2} \frac{t_0}{\tau} \eta = \frac{c_0^2}{c_\infty^2} \frac{t}{\tau}$$

and involving expansion (7) for $c_{\infty} \gg c_0$. Then we have

using (12) at T=0

$$S_{\tau}(0) = S_0 \frac{c_0^2}{c_\infty^2} \frac{t_0}{\tau} s_{\tau}[\boldsymbol{v}(\boldsymbol{x}, \eta)]$$
 (15)

where the dimensionless action $s_{\tau}[\boldsymbol{v}(\boldsymbol{x}, \eta)]$ reads

$$s_{\tau}[\boldsymbol{v}(\boldsymbol{x},\,\eta)] = \frac{1}{4\pi} \int_{-\infty}^{\infty} d\eta d\eta' \int d^3x \frac{(\boldsymbol{v}(\boldsymbol{x},\eta) - \boldsymbol{v}(\boldsymbol{x},\eta'))^2}{(\eta - \eta')^2} + \int_{-\infty}^{\infty} d\eta \int d^3x \left(\frac{1}{2}(\nabla\chi)^2 + \frac{1}{2}\chi^2 + \frac{1}{3}\chi^3\right)$$

Thus, as the relaxation time increases, the extremum value of effective action decreases gradually and reduces by factor c_0/c_∞ with saturation at $\tau = \infty$. In the intermediate region $t_0c_0^2/c_\infty^2 \ll \tau \ll t_0c_0/c_\infty$ the action falls approximately as $1/\tau$.

Analyzing temperature behavior of the effective action, of course one should take into account the temperature dependence of the thermodynamic quantities involved into the nucleation process. However, most important aspect of the temperature behavior for the nucleation probability is associated with the temperature dependence of relaxation time $\tau = \tau(T)$. It is obvious that one should compare the relaxation time $\tau(T)$ with the typical time of the order of t_0 necessary for the underbarrier growth of a nucleus. Involving the frequency dispersion in (7), we introduce two temperatures $T_{\tau 1}$ and $T_{\tau 2}$ defined according to

$$\tau(T_{\tau 1}) \sim \frac{c_0}{c_\infty} t_0 = \frac{\sqrt{\lambda \rho}}{c_0 c_\infty}$$

$$\tau(T_{\tau 2}) \sim \frac{c_0^2}{c_\infty^2} t_0 = \frac{\sqrt{\lambda \rho}}{c_\infty^2}$$
(16)

Keeping in mind that usually the relaxation time does not diminish as the temperature lowers, we thus suppose $T_{\tau 1} < T_{\tau 2}$. Note that these parameters enter naturally Eq. (13) and Eq. (14), determining the order-of-magnitude correction comparable with action S_0 and S_{∞} , respectively. The physical meaning of temperatures $T_{\tau 1}$ and $T_{\tau 2}$ lies in separating the high frequency type of underbarrier nucleation processes at $T \ll T_{\tau 1}$ from the low frequency type at $T \gg T_{\tau 2}$. The magnitude of the transient region depends on a ratio c_{∞}/c_0 .

Let us now turn to the estimate of the thermalquantum crossover temperature T_q . At first, we assume that T_q is above $T_{\tau 2}$. Then, using $S_0(0)$, we can estimate the thermal-quantum crossover temperature approximately as

$$T_q \sim T_{q\,0} \approx \frac{U_0}{S_0(0)} = \frac{f_0}{s_0} \frac{1}{t_0} = 0.27 \frac{c_0^2}{\sqrt{\lambda \rho}} , \text{ if } t_0 T_{\tau 2} \ll 1$$
 (17)

In the opposite case provided T_q is below $T_{\tau 1}$ we should employ S_{∞} . This yields

$$T_q \sim T_{q\,\infty} = \frac{c_\infty}{c_0} T_{q\,0} \;, \; \text{if} \; \frac{c_0}{c_\infty} t_0 T_{\tau 1} \gg 1 \qquad (18)$$
 In the intermediate case, assuming $T_{\tau 1} < T_q < T_{\tau 2}$, we

In the intermediate case, assuming $T_{\tau 1} < T_q < T_{\tau 2}$, we should use Eq. (15) to estimate the thermal-quantum crossover temperature. Then we find

$$T_q \sim \frac{c_\infty^2}{c_0^2} \frac{\tau}{t_0} T_{q0}$$
, if $t_0 T_{\tau 1} \ll \frac{c_\infty^2}{c_0^2} \frac{\tau}{t_0} \ll t_0 T_{\tau 2}$ (19)

Thus, the larger the relaxation time, the higher the crossover temperature.

In order to treat the thermal-quantum crossover more detailed, we consider the stability of the classical path $\mathbf{v}_{\rm cl}(\mathbf{x})$ with respect to its small perturbations $\delta \mathbf{v}(\mathbf{x}, \eta)$, representing an arbitrary path as $\mathbf{v}(\mathbf{x}, \eta) = \mathbf{v}_{\rm cl}(\mathbf{x}) + \delta \mathbf{v}(\mathbf{x}, \eta)$ and, correspondingly, $\chi(\mathbf{x}, \eta) = \chi_{\rm cl}(\mathbf{x}, \eta) + \delta \chi(\mathbf{x}, \eta)$. Then we have, being $\chi = -\nabla \cdot \mathbf{v}$,

$$s[\delta \boldsymbol{v}(\boldsymbol{x}, \eta)] = \frac{f_0}{T'} + \int_{-1/2T'}^{1/2T'} d\eta d\eta' \int d^3x \, \frac{1}{2} \delta \dot{\boldsymbol{v}}(\boldsymbol{x}, \eta) D(\eta - \eta') \delta \dot{\boldsymbol{v}}(\boldsymbol{x}, \eta')$$
$$+ \int_{-1/2T'}^{1/2T'} d\eta \int d^3x \, \left[\frac{1}{2} (\nabla \delta \chi)^2 + (1 + 2\chi_{\text{cl}}(\boldsymbol{x})) \frac{\delta \chi^2}{2} + \frac{\delta \chi^3}{3} \right]$$

To find the point of instability in temperature, it is sufficient to retain quadratic terms alone. Turning to Fourier representation in time, we arrive at the expansion

$$\delta \boldsymbol{v}(\boldsymbol{x}, \eta) = T' \sum_{n} \delta \boldsymbol{v}_{n}(\boldsymbol{x}) e^{-\imath \Omega_{n} \eta}, \ \delta \boldsymbol{v}_{-n} = \delta \boldsymbol{v}_{n}^{*}$$

$$s[\delta \boldsymbol{v}_n(\boldsymbol{x})] = \frac{f_0}{T'} + \int d^3x \left\{ \frac{1}{2} T' \sum_n |\Omega_n|^2 D(\Omega_n) \delta \boldsymbol{v}_n \delta \boldsymbol{v}_{-n} + \nabla \delta \chi_n \cdot \nabla \delta \chi_{-n} + (1 + 2\chi_{\text{cl}}(\boldsymbol{x})) \delta \chi_n \delta \chi_{-n} + \ldots \right\}$$

Next, we decompose an arbitrary perturbation $\delta v_n(x)$ into a series

$$\delta oldsymbol{v}_n(oldsymbol{x}) = \sum_{lpha} C_{n,\,lpha} oldsymbol{w}_lpha(oldsymbol{x})$$

over a complete orthonormal set of eigenfunctions \boldsymbol{w}_{α} of the equation

$$\nabla[(-\nabla^2 + (1 + 2\chi_{\rm cl}(\boldsymbol{x}))(-\nabla \cdot \boldsymbol{w})] = E\boldsymbol{w}$$
 (20)

Then one has retaining quadratic terms alone

$$s[C_{n,\alpha}] = \frac{f_0}{T'} + \frac{T'}{2} \sum_{n,\alpha} \{ |\Omega_n|^2 D(\Omega_n) + E_{\alpha} \} |C_{n,\alpha}|^2$$

where E_{α} is the eigenvalues of Eq. (20). Provided the

expression in braces becomes negative, at least, for one mode, the classical path becomes absolutely unstable and the crossover to the quantum path dependent on time is unavoidable. As the temperature lowers, the mode which first becomes unstable is a mode with n=1 and with $\alpha=1$ corresponding to the minimum negative value of E_{α} . Thus the temperature T_1 of absolute instability of the classical path is determined by

$$|\Omega_1|^2 D(\Omega_1) = |\Omega_1|^2 \frac{1 + |\Omega_1| \tau/t_0}{1 + |\Omega_1| \tau c_{\infty}^2/t_0 c_0^2} = -E_1$$

Solving equation yields

$$T_{1} = \begin{cases} \frac{\sqrt{|E_{1}|}}{2\pi} \frac{1}{t_{0}} \left(1 + \frac{c_{\infty}^{2} - c_{0}^{2}}{c_{0}^{2}} \frac{\tau}{t_{0}} \sqrt{|E_{1}|} + \ldots \right), & \text{if} & \frac{t_{0}}{\tau(T_{1})} \gg \frac{c_{\infty}^{2}}{c_{0}^{2}} \sqrt{|E_{1}|} \\ \frac{|E_{1}|}{2\pi} \frac{\tau}{t_{0}^{2}} \frac{c_{\infty}^{2}}{c_{0}^{2}}, & \text{if} & \frac{c_{\infty}^{2}}{c_{0}^{2}} \sqrt{|E_{1}|} \gg \frac{t_{0}}{\tau(T_{1})} \gg \frac{c_{\infty}}{c_{0}} \sqrt{|E_{1}|} \\ \frac{c_{\infty}}{c_{0}} \frac{\sqrt{|E_{1}|}}{2\pi} \frac{1}{t_{0}} \left(1 - \frac{1}{2} \frac{c_{\infty}^{2} - c_{0}^{2}}{c_{\infty}^{2}} \frac{c_{0}}{c_{\infty}} \sqrt{|E_{1}|} \frac{t_{0}}{\tau} + \ldots \right), & \text{if} & \frac{c_{\infty}}{c_{0}} \sqrt{|E_{1}|} \gg \frac{t_{0}}{\tau(T_{1})} \end{cases}$$

$$(21)$$

Value E_1 can also be found with the help of the variational principle, minimizing the functional

$$E = \frac{\int d^3x \left[(\nabla \chi)^2 + (1 + 2\chi_{\rm cl}(\boldsymbol{x}))\chi^2 \right]}{\int d^3x \, \boldsymbol{w}^2} \quad , \quad \chi = -\nabla \cdot \boldsymbol{w}$$

The approximate value $E_1 \approx -6.11$ yields temperature $T_1 \sim 0.39/t_0$ correcting the estimate $T_{q0} \sim 0.27/t_0$ for the case $\tau = 0$ in (17). As is seen from (21), the calculations of temperature T_1 for absolute instability of the classical path are in agreement with the estimates in (17)–(19). The temperature of the thermal-quantum crossover T_q cannot be lower than T_1 .

5. DISCUSSION

Discussing common implications for quantum decay of a metastable liquid, first of all we should emphasize that the relaxation time is an important parameter governing the nucleation process. The quantum nucleation rate proves to be a monotone increasing function of relaxation time τ , saturating in the limit of infinite relaxation time $\tau=\infty$. Depending on a ratio of high frequency sound velocity to low frequency one c_{∞}/c_0 , we can distinguish either two or three quantum nucleation regime. If $c_{\infty}/c_0 \gtrsim 1$, we have the low frequency or high frequency regime depending on the relationship between the relaxation time and the vicinity to the spinodal. In the case of strong inequality $c_{\infty}/c_0 \gg 1$ it becomes possible to discern the crossover between the low and high frequency regimes as an independent regime corresponding to the overdamped viscous quantum nucleation.

Second, provided the relaxation time $\tau(T)$ as a function of temperature diverges for $T \to 0$, the nucleation rate W = W(T) can demonstrate a non-monotone temperature behavior with a minimum in the region of the thermal-quantum crossover temperature T_q . Accordingly, if the nucleation rate is fixed under experimental conditions, the observable supersaturation of the metastable phase will be maximum. The larger the ratio c_{∞}/c_0 , the more prominent the relative magnitude of the

effect.

The third aspect of nucleation concerns the behavior the thermal-quantum crossover temperature as one approaches the spinodal point where the sound velocity vanishes $c_0(\rho_c)=0$. Keeping in mind $\tau^{-1}(T\to 0)\to 0$, one can see that the crossover temperature T_q reduces in a linear proportion to the product $c_\infty(\rho)c_0(\rho)$ vanishing at the spinodal point $\rho=\rho_c$ together with the potential barrier. Thus the nucleation process in the region of metastability immediately adjacent to the spinodal should be governed by the classical thermal activation.

Let us compare liquid ³He and ⁴He. In Bose-liquid ⁴He the thermal-quantum crossover temperature is not expected to exceed approximately 1K. For definiteness, we consider the temperature region below 0.6K when the contribution from rotons into all phenomena becomes insignificant and excitations in liquid can be treated as a purely phonon gas. Since the process of sound propagation in ⁴He is closely connected with the propagation of phonons, the velocities of both low frequency and high frequency sound do not differ much from the phase velocity c of phonons. The deviation from the velocity of phonons is wholly due to the presence of thermal excitations with normal density $\rho_n = 2\pi^2 T^4/(45\hbar^3 c^5)$. The difference in velocities c_{∞} and c_0 can be found using, e.g., [23]

$$c_{\infty} - c_0 = c \frac{\rho_n(T)}{\rho} \left[\frac{3}{4} (A+1)^2 \log \frac{1}{\gamma} \left(\frac{c}{2\pi T} \right)^2 - \frac{(3A+1)^2}{4} - 3A - 2 \right]$$

Here $A=\partial \log c/\partial \log \rho$ is the Grüneisen parameter and γ is the coefficient of the cubic term describing deviation of the phonon dispersion from the linear one. The relaxation time in the system of phonons grows drastically at low temperatures

$$\frac{1}{\tau} \sim (A+1)^4 \frac{\hbar^2}{\rho^2 c} \left(\frac{T}{2\pi\hbar c}\right)^9$$

Such large relaxation time at $T \to 0$ results in temperature $T_{\tau 1}$ always higher than the thermal-quantum crossover temperature T_q . In the vicinity of the spinodal one has $T_{\tau 1} \propto c^{8/9}$ and crossover temperature $T_q \propto c_0 c_\infty \sim c^2$. Thus the regime of quantum nucleation corresponds to the high frequency limit and the underbarrier growth of a nucleus occurs under collisionless conditions, i.e., typical size of a nucleus is much less compared with the mean free path of excitations. The latter favors the quantum nucleation.

However, the temperature effect associated with the distinction in velocities c_{∞} and c_0 and also with their temperature dependence, which becomes of the order of 1ppm below 0.1K, is not large. The relative contribution into the effective action has the order of the magnitude of small parameter ρ_n/ρ . In addition, though the low frequency and high frequency regimes of quantum

nucleation differs significantly in kind from the physical point of view, the quantitative difference between the regimes is again moderate since the relative change of the sound velocity from the low to the high frequency limit is about the same small ratio $(c_{\infty}-c_0)/c\sim\rho_n/\rho$. From the experimental point of view this can hardly be discerned unless a rather precise measurement of the nucleation rate is employed. On the whole one should not expect any noticeable temperature variations in the rate of quantum nucleation at low temperatures. Apparently, such picture takes place in the experiments on nucleation of solids or cavities in superfluid ⁴He where the temperature-independent nucleation rate is observed at temperatures below one or more hundred of mK.

Let us turn now to normal liquid 3 He. In Fermi liquid the nature of the low and high frequency sound modes, associated with the various physical mechanisms, differs in a qualitative sense. Thus, unlike superfluid 4 He, sound velocities c_0 and c_∞ are different even at zero temperature. The relative difference amounts about 4% at zero pressure. The relaxation time due to collisions between 3 He quasiparticle excitations varies with temperature as

$$\tau = \nu/T^2 \quad (\nu \approx 2 \cdot 10^{-12} \ s \cdot K^2)$$
 (22)

Generally speaking, the numerical coefficient depends on pressure or density of a liquid.

Within the framework of the Fermi liquid theory the instability at spinodal $c_0(\rho_c)=0$ implies for the Landau parameter $F_0=-1$. However, velocity c_∞ of the high frequency collisionless mode may remain finite since the condition of the thermodynamic stability $c_0^2>0$ cannot be applied for nonequilibrium processes. Unlike the usual case of $F_0>0$, the existence of high frequency zero-sound mode depends on the magnitude of the next Landau parameter F_1 responsible for the value of the effective mass and Fermi velocity of quasiparticles. The solution of the dispersion equation for zero-sound mode [23]

$$\frac{s}{2}\log\frac{s+1}{s-1} - 1 = \frac{1 + F_1/3}{F_0(1 + F_1/3) + F_1s^2}$$

where $s=c_{\infty}/v_F$ is a ratio of the velocity of zero-sound wave to the Fermi velocity, remains real at $F_0=-1$ provided $F_1>3/2$. To estimate F_1 at the spinodal, we use an extrapolation [24] of the dependence of the effective mass on density into the negative pressure region from the fit of the data at positive pressures

$$(1 + F_1/3)^{-1} = m/m^* = [1.0166(1 - 5.138\rho)]^2$$

Substituting spinodal density $\rho_c = 0.054$ g/cm³ as estimated in [24], we have approximately $F_1 \approx 3$ and arrive at zero-sound velocity almost same as the Fermi one s = 1.006 instead of $F_1 = 6.25$ and s = 3.6 at zero pressure. On the whole, reduction of the effective mass prevails over reduction of the density and should result even in increasing the Fermi velocity by approximately

one-third as compared with the case of zero pressure. Eventually, the only essential point is that the high and low frequency sound velocities are expected to have various limiting behavior at negative pressures and their ratio c_{∞}/c_0 enhances infinitely with approaching the spinodal.

Let us introduce temperature T_{ν} at which the thermal energy of excitations is about of quantum uncertainty in energy due to collisions between excitations, i.e.,

$$\tau(T_{\nu}) = 1/T_{\nu}$$

In essence, this temperature determines the upper limit of applicability of the Fermi-liquid theory. According to (22) $T_{\nu} = \nu \approx 0.26$ K and is well below the Fermi temperature. The next speculation depends the dimensionless parameter

$$\zeta = \nu \tau(T_{\tau 2}) = \frac{\nu \sqrt{\lambda \rho}}{c_{\infty}^2}$$

Comparing temperatures T_{ν} and $T_{\tau 2}$ defined in (16), we find the relationship

$$T_{\tau 2} = T_{\nu} \zeta^{-1/2}$$

At first, we consider the most diversified case $\zeta \ll 1$ when $T_{\tau 2} > T_{\nu}$. In accordance with (21) we can discern three regions in the behavior of the thermal-quantum crossover temperature as a function of the vicinity to the spinodal point

$$T_q \simeq \begin{cases} \frac{c_{\infty}}{c_0} \frac{\sqrt{|E_1|}}{2\pi} \frac{1}{t_0} \propto c_0 , & \text{if} \quad c_0 < c_{\infty} \zeta \\ \left(\frac{|E_1|}{2\pi} \frac{\nu}{t_0^2} \frac{c_{\infty}^2}{c_0^2} \right)^{1/3} \propto c_0^{2/3} , & \text{if} \quad c_{\infty} \zeta < c_0 < c_{\infty} \zeta^{1/4} \\ \\ \frac{\sqrt{|E_1|}}{2\pi} \frac{1}{t_0} \propto c_0^2 , & \text{if} \quad c_{\infty} \zeta^{1/4} < c_0 \end{cases}$$

Thus, depending on the relation between ζ and c_0/c_∞ , we can observe either one, or two, or three types of nucleation in the quantum region as the temperature varies from zero one to the thermal-quantum crossover. While $c_0/c_\infty < \zeta$ one has only the high-frequency collisionless type. Within intermediate region $\zeta < c_0/c_\infty < \zeta^{1/4}$ there is a crossover to the overdamped type at $T \sim T_{\tau 1}$ and at $T \sim T_{\tau 2}$, if $c_0/c_\infty > \zeta^{1/4}$, the nucleation process becomes a low-frequency collisional one. Compared with the zero temperature value the effective action increases with the temperature as T^2 due to enhancement of the collision frequency $\tau^{-1}(T)$. Correspondingly, the nucleation rate decreases. The relative effect at the thermal-quantum crossover temperature when the nucleation rate is minimal is most prominent if $c_0/c_\infty > \zeta^{1/4}$.

The case when $\zeta>1$ is meagre. Here, since always $c_0<\zeta c_\infty$, in the quantum regime we can have only the high-frequency collisionless regime with the next crossover to the thermal Arrhenius nucleation. The temperature behavior of the effective action and nucleation

rate demonstrates the same features as in the above-considered case $\zeta \ll 1$ though not so well-marked.

Concerning the numerical estimate of parameter ζ , one should evaluate, first of all, parameter λ related closely with the spatial dispersion of sound in the long-wavelength limit. Thus we need in the reliable estimate of λ near the spinodal point. As a rule, the spatial dispersion, associated with the interatomic spacing a, becomes significant at wave vectors close to $k \sim 1/a$. To estimate the order of the magnitude for λ , we put that nonlinear term in the sound dispersion due to $\lambda(\nabla \rho)^2$ becomes comparable with the main linear term at $k \sim 1/a$. Since we suppose that λ remains finite at the spinodal, it is convenient to represent

$$\lambda \rho \simeq c_{\infty}^2 a^2$$

entailing $\zeta \simeq \nu a/c_{\infty}$. Involving that the Fermi momentum is also about 1/a and c_{∞} is comparable with the Fermi velocity, we arrive at the dimensional estimate for parameter ζ

$$\zeta \sim T_{\nu}/T_F$$

So, in liquid ³He one may expect that $\zeta \lesssim 1$, entailing a noticeable minimum in the temperature behavior of the nucleation rate in thermal-quantum crossover region. Provided the nucleation rate keeps constant in experiment, this means that attainable deviation $\delta \rho = \rho - \rho_c$ and, correspondingly, negative cavitation pressure P pass through a minimum in the course of the crossover from the thermal-to-quantum regime with the next almost temperature-independent behavior at sufficiently lower temperatures. The similar temperature behavior of the supersaturation due to the effect of relaxation processes on the quantum nucleation is observed in supersaturated ³He-⁴He liquid mixtures [6]. As it concerns the cavitation experiments in ³He [11], in this sense the observation of a sharp increase of the cavitation threshold at temperatures below about 60 mK can serve for some evidence of the incipient thermal-quantum crossover.

In conclusion, we have suggested a theory which, for the first time, involves the relaxation and high frequency properties of a condensed medium into quantum decay of a metastable liquid near the spinodal at low temperatures. The model developed can be employed for clarifying the physical picture of the low temperature cavitation in liquid ³He and ⁴He at negative pressures. The results obtained might thus be a helpful guide for experiments on the quantum decay in a metastable condensed medium.

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